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**Movement of Outdoor Particles to
the Indoor Environment: An Analysis of
the Arnhem Lead Study**

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INTRODUCTION

Particles in the indoor environment are derived from various indoor and outdoor sources. Historically, much research has focused on the generation of airborne particles from combustion sources outdoors (e.g., sulfates and lead) or indoors (e.g., cigarette smoke) and the associated levels in indoor air, as measured by fixed or personal air samplers.^{1,2,3} Another potential source of indoor particles that has not received as much attention is soil that is tracked into homes by residents and resuspended into indoor air by walking, vacuuming, etc. This transport process is of potential concern because it brings soil-derived contaminants into the indoor environment where individuals can contact the contaminated dust/soil via incidental ingestion, dermal contact, and direct inhalation. These exposure mechanisms could be just as important as direct contacts with contaminated soil outdoors because people spend most of their time indoors while at home.⁴ The goal of this paper is to analyze the role of soil tracking as a source of indoor particles and to quantify key parameters influencing the transport of soil-derived particles, namely resuspension rates for particulate matter on floors and the deposition velocities of suspended particles originating in indoor and outdoor air. We begin the paper with a brief review of studies that address pertinent aspects of particle transport processes and present a simple model for studying the transport of particles in the indoor environment. We then use the model to examine data on the distributions of Pb in the indoor and outdoor environments of a community situated adjacent to a secondary lead smelter.

BACKGROUND

Source-term analyses of suspended particles in indoor air have often attributed a portion of the airborne particles indoors to personal activities such as vacuuming, cooking, and walking. The true importance of indoor sources, though, has been somewhat obscured by the assumption that the concentrations of outdoor particles are reduced indoors by the filtering effect of building shells.¹ In addition, little information is available on the rates with which particles are suspended into indoor air as a result of human activities. Recent studies, though, have provided new information on the factors influencing indoor/outdoor relationships of particles and the resuspension of particles from floor surfaces.

Filtration of particles by the building shell

Concentrations of airborne particles indoors that are lower than outdoor levels have been explained by the filtration loss of airborne particles as they are drawn through the building shell by air exchange. The filtration of particles in the absence of indoor sources has been modeled using a penetration factor, P , defined as

$$P = \left[\frac{C_{in}}{C_{out}} \right] \frac{(\lambda_d + Ach)}{Ach}, \quad (1)$$

where

C_{in} = average concentration of a contaminant in suspended particles in indoor air, $\mu\text{g m}^{-3}$;

- A_{ch} = air exchange rate, h^{-1} ;
 λ_d = deposition loss rate of particles, h^{-1} , and
 C_{out} = average concentration of a contaminant in suspended particles in outdoor air, $\mu g\ m^{-3}$.

When the loss rate of suspended particles is small compared to the air exchange rate, the value of P is simply equal to the ratio of the concentration of a contaminant in indoor air to its concentration in outdoor when no indoor sources are present. In order to evaluate the value of P , measurements are required of the indoor/outdoor levels of particles of a given size class, as well as the deposition loss rates for the particles and the air exchange rate. Thatcher and Layton⁵ instrumented a two-story, single-family, detached residence in Livermore, California, during the summer of 1993 to quantify the input parameters required to estimate the amount of particle penetration through the house shell. The deposition velocities for coarse particles (1 to 5 μm , 5 to 10 μm , and 10 to 25 μm optical particle diameters) in the residence were determined by raising the indoor particle concentration and monitoring the particle loss rates and air-infiltration rate over time. To determine the penetration factor, indoor and outdoor particle concentrations were measured while the residence was unoccupied, minimizing the effect of resuspension. They found that the value of P is essentially unity for particles ranging in size from 1 to 25 μm in optical diameter. The reduced concentrations of particles measured indoors were explained when the deposition loss was included in Eq. 1. Although building practices that lead to building shells that are tighter than the one studied might be expected to produce values of P that are less than one, this would also lead to a reduction in the infiltration rate and a corresponding reduction in the importance of infiltrating particles in the overall mass balance. Therefore, even in very tight buildings, particles losses due to gravitational settling would still be expected to be much greater than losses of infiltrating particles.

Resuspension from floors

Few direct measurements are available on the rates with which particles on floors are suspended into air (expressed as a fraction of particulate loading on floor surfaces suspended per unit time) by human activities. Healy⁶ cited studies that showed that the resuspension rate can exceed $1 \times 10^{-3}\ h^{-1}$ for particles on non-carpeted surfaces. Based on a review of resuspension data and assumptions regarding the amounts of time spent at different activities indoors, Healy estimated a time-weighted-average resuspension rate of $5 \times 10^{-4}\ h^{-1}$ for a house. Murphy and Yocom⁷ selected a value of $10^{-4}\ h^{-1}$ for use in an analysis of the sources of particles in the indoor environment. Thatcher and Layton⁵ measured the increased concentrations of suspended material from floor surfaces in a house and found that resuspension decreases as the optical particle diameter decreases, with an apparent resuspension threshold of about 2 μm . Kamens et al.⁸ found a similar trend between optical particle size and particle levels indoors associated with varying patterns of human activity. Based on these findings, fine particles indoors (i.e., less than about 2 μm in diameter) should not be influenced strongly by indoor activities. And as a result, concentrations of fine particles indoors (and associated contaminant species) would be expected to have a higher correlation coefficient with outdoor levels than would coarse particles, which are more susceptible to resuspension indoors. The experimental results of Clayton et al.² are consistent with this relationship. They reported a correlation coefficient of 0.70 between the concentration of fine particulate matter (i.e., denoted $PM_{2.5}$ for particles

under 2.5 μm in diameter) in the main living area of a sample of houses and the outdoor levels recorded at a fixed monitor. In contrast, PM_{10} concentrations had a correlation coefficient of 0.51, while concentrations obtained from personal air samplers had a correlation coefficient of only 0.37 with the fixed-site monitor. The low correlation coefficient for personal-air samplers suggests that a large portion of the particles collected were suspendable particles over 2 μm in diameter.

BASIC MODEL

We have formulated a simple steady-state model that predicts the concentration of Pb in indoor air as the sum of the contributions from resuspension from floors/carpets and infiltration from outdoor air contaminated by emissions from a secondary lead smelter operating with unspecified pollution controls. The model as currently formulated is based on the assumption that both resuspended and outdoor Pb particles consist primarily of coarse-mode particulates (i.e., $\geq 2 \mu\text{m}$ in diameter) that deposit on horizontal indoor surfaces by gravitational settling. Separate deposition velocities are used for the floor- and outdoor-derived particles to reflect differences in the particle sizes associated with the two sources, although we assume that the deposition surface areas are equal. The model is expressed as

$$C_{\text{in}} = \frac{L_{\text{fl}}R}{(\bar{v}(\text{c})_{\text{d}} + \text{Ach} \cdot h)} + \frac{\text{Ach}C_{\text{out}}h}{(\bar{v}(\text{o})_{\text{d}} + \text{Ach} \cdot h)}, \quad (2)$$

where

- C_{in} = concentration of Pb in indoor air, $\mu\text{g m}^{-3}$;
- C_{out} = concentration of Pb in outdoor air, $\mu\text{g m}^{-3}$;
- L_{fl} = mass loading of lead on accessible floor surfaces, $\mu\text{g m}^{-2}$;
- R = resuspension rate of coarse particles from floors, h^{-1} ;
- h = ceiling height, m (assumed to equal 2.4 m);
- $\bar{v}(\text{c})_{\text{d}}$ = average deposition velocity of resuspended particles, m h^{-1} , and
- $\bar{v}(\text{o})_{\text{d}}$ = average deposition velocity of outdoor particles, m h^{-1} .

Data on the concentrations of lead in particulate matter in outdoor air, indoor air, and on floors, can be used to define the following mass-balance relationship

$$C_{\text{p}}^{\text{in}} = aC_{\text{fl}} + (1 - a)C_{\text{p}}^{\text{out}}, \quad (3)$$

where

- a = fraction of lead in suspended indoor particles derived from floor particles ($0 \leq a \leq 1$), unitless;
- C_{p}^{in} = concentration of lead in suspended particles in indoor air, μgg^{-1} ;
- C_{fl} = concentration of lead in particulate matter on floors, μgg^{-1} ; and
- $C_{\text{p}}^{\text{out}}$ = concentration of lead in suspended particles in outdoor air, μgg^{-1} .

Given that $C_{in} = C_p^{in}TSP_{in}$, $L_{fl} = C_{fl}M_{fl}$, and $C_{out} = C_p^{out}TSP_{out}$, we obtain the following equations

$$a = \frac{\frac{M_{fl}R}{(\bar{v}(c)_d + Ach \cdot h)}}{TSP_{in}} = \frac{(C_p^{in} - C_p^{out})}{(C_{fl} - C_p^{out})} \quad (4)$$

and

$$1 - a = \frac{\frac{AchTSP_{out}h}{(\bar{v}(o)_d + Ach \cdot h)}}{TSP_{in}} = 1 - \frac{(C_p^{in} - C_p^{out})}{(C_{fl} - C_p^{out})}, \quad (5)$$

where

- TSP_{in} = loading of particulate matter in indoor air, $g\ m^{-3}$;
- M_{fl} = loading of particulate matter on floor surfaces, $g\ m^{-2}$; and
- TSP_{out} = loading of particulate matter in outdoor air, $g\ m^{-3}$.

A similar mass-balance relationship can be developed for the average concentration of lead in dustfall on horizontal indoor surfaces, that is,

$$C_{df} = bC_{fl} + (1 - b)C_p^{out}, \quad (6)$$

where

- C_{df} = concentration of a contaminant in dustfall on indoor surfaces, $\mu g\ g^{-1}$; and
- b = fraction of contaminant in dustfall derived from floor particles ($0 \leq b \leq 1$), unitless.

The contributions of the two sources of lead-bearing particles is a function of the average deposition velocities of the outdoor and floor-derived particles suspended in indoor air. The fractional contributions can be estimated using the total average deposition rate of particles onto horizontal surfaces, denoted F_{df} , or

$$b = \frac{\left[\frac{M_{fl}R}{(\bar{v}(c)_d + Ach \cdot h)} \right] \bar{v}(c)_d}{F_{df}} = \frac{(C_{df} - C_p^{out})}{(C_{fl} - C_p^{out})} \quad (7)$$

$$1 - b = \frac{\left[\frac{\text{AchTSP}_{\text{out}} h}{(\bar{v}(o)_d + \text{Ach} \cdot h)} \right] \bar{v}(o)_d}{F_{\text{df}}} = 1 - \frac{(C_{\text{df}} - C_p^{\text{out}})}{(C_{\text{fl}} - C_p^{\text{out}})} \quad (8)$$

where F_{df} has units of $\text{g m}^{-2} \text{h}^{-1}$. The paired equations 4 and 7 and 5 and 8 (for resuspended particles and outdoor-derived particles, respectively) can be solved simultaneously using the measured input parameters (summarized in Table 1) to determine the values of Ach, $\bar{v}(c)_d$, $\bar{v}(o)_d$, and R.

ANALYSIS OF THE ARNHEM DATA

The Arnhem Lead Study³ consisted of a comprehensive investigation in 1978 of the levels of lead and particulate matter in the indoor and outdoor environment of the city of Arnhem, The Netherlands. The principal source of airborne lead was a secondary lead smelter. As part of the overall study, measurements were made of suspended particulate matter indoors and outdoors, the loading of particles on floor surfaces, dustfall of indoor particles, and the concentrations of lead in those media. Samples of lead in particulate matter were collected from over 100 houses that were located at distances of 450 to 1000 m from the lead smelter. The concentrations of Pb in the particles sampled ranged from $6400 \mu\text{g g}^{-1}$ for ambient airborne particles derived from the secondary lead smelter to $280 \mu\text{g g}^{-1}$ for coarse particles on floor surfaces (see Fig. 1). The Pb content of suspended particles indoors was $2200 \mu\text{g g}^{-1}$ and in dustfall it was $1000 \mu\text{g g}^{-1}$. The decrease in the Pb levels in particulate matter indoors from the levels in suspended particles outdoors suggests that particulate Pb indoors was diluted by the resuspension of coarse particulate matter on floors containing low Pb levels. The mass loading of particulate matter on floors was 0.26 g m^{-2} and the average concentration of Pb in floor dust was $480 \mu\text{g g}^{-1}$. The concentration of Pb in fine dust particles (i.e., particles retained on glass-fiber filters of 150-mm diameter) was $960 \mu\text{g g}^{-1}$ and $280 \mu\text{g g}^{-1}$ in coarse dust particles. Thus, nearly 70% of the Pb mass on floors was associated with coarse particles (0.18 g m^{-2}) and the remainder was associated with fine particles. The coarse particles are presumed to come from the tracking of outdoor soils (Pb concentration of $240 \mu\text{g g}^{-1}$) to floor surfaces. If we assume that the coarse fraction best represents the suspendable mass of particles on floors, based on the experimental studies described above, then we can use Eqs. 3 and 6 to estimate the contributions of suspended outdoor particles and floor dust to the average concentrations of Pb in suspended indoor particles and dustfall. Figure 2 shows the relative contributions of the two Pb sources to Pb concentrations in indoor particles. In both cases, floor-derived Pb is the dominant source of Pb in suspended particles and dustfall indoors.

The reduction of Pb in dustfall by a factor of about two from the concentration in total suspended particles, due to resuspension from floors, is potentially important from an exposure standpoint because such particles deposit on horizontal surfaces (furniture, tables, beds, etc.) that adults and children come in contact with. Consequently, such surfaces should be distinguished from other contact media (e.g., dust on floors, soils, etc.) in epidemiological studies attempting to relate environmental Pb levels with levels of Pb in blood in exposed populations.

Using the estimates of a and b from Eqs. 3 and 6 along with the other input parameters presented in Table 1 and Eqs. 5 and 8, we estimated the air exchange rate (Ach) to be 0.6 h^{-1} , a rate that is in the middle of the range defined by housing with low air exchange ($\sim 0.2 \text{ h}^{-1}$) and with elevated air exchange ($\sim 1 \text{ h}^{-1}$). The estimated settling velocity of the Pb-bearing particles from outdoor air ($\bar{v}(o)_d$) is 1 m h^{-1} . This settling velocity corresponds to particles with diameters of 2.2 and $2.5 \mu\text{m}$, respectively, assuming particle densities of 2 and 1.5 g cm^{-3} . For comparison, the settling velocity for indoor Pb particles obtained from measurements of the Pb settling rate and the concentration in indoor air was 1.2 m h^{-1} (i.e., $0.31 \mu\text{g m}^{-2} \text{ h}^{-1} \div 0.26 \mu\text{g m}^{-3}$). We solved Eqs. 4 and 8 simultaneously to determine an average settling velocity for coarse particles ($\bar{v}(c)_d$) of 3 m h^{-1} (corresponding to particle diameters of 3.7 and $4.3 \mu\text{m}$ for particle densities of 2 and 1.5 g cm^{-3}). The settling velocity for suspended particles calculated from measured fluxes and the TSP concentration indoors was 2.7 m h^{-1} (i.e., $0.00032 \text{ gm}^{-2} \text{ h}^{-1} \div 0.00012 \text{ gm}^{-3}$). The solution for the resuspension rate (R) yields a value 0.002 h^{-1} . Although this value is a factor of four higher than that the rate predicted by Healy (i.e., 0.0005 h^{-1}) on the basis of an assumed mix of human activities, we note that Diemel et al. reported that the occupants of the study houses vacuumed nearly every day, thereby increasing the transfer of floor particles to indoor air.

CONCLUSIONS

The Arnhem Lead Study provides a unique set of measurements for studying the relationship between Pb in the outdoor environment near a point source (i.e., a secondary lead smelter) and in the indoor environment of nearby homes. Our analysis of the data collected as part of that study indicates that the movement of Pb-bearing soils into the houses by tracking, followed by resuspension by human activities such as walking and vacuuming accounts for a significant portion of the Pb in indoor particulate matter. Although the concentration of airborne lead in ambient particles outdoors was much higher than the concentration of Pb in floor dust, the suspension of particulate matter from floors essentially diluted the quantity of airborne lead derived from infiltrating outdoor air. Our estimate of the settling velocities for both resuspended and outdoor particles are consistent with the nature of the Pb-bearing particles generated by the different sources.

The relationship between contaminants in outdoor soil and the associated levels in the indoor environment requires additional studies to clarify the role of human factors (e.g., occupancy, cleaning frequencies, etc.) and housing factors (e.g., carpeted surfaces, type of vacuum cleaner, filtration by heating/cooling equipment etc.) in modifying indoor levels. A related issue involves the selection of the most appropriate sampling strategies to characterize transport processes affecting the movement of soil-derived contaminants to and within the indoor environment. Fortunately, the Arnhem Lead Study included enough measurements of Pb-bearing particles that we were able to derive important variables such as the air exchange rate and deposition velocities for indoor particles. Future studies are needed that address specifically a set of parameters that are linked to models that describe the transport processes influencing the migration of soil contaminants to the indoor environment.

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Table 1. Input parameters derived from the Arnhem Lead Study. Values are presented to two significant figures.

Parameter	Units	Value
TSP _{in}	g m ⁻³	1.2×10^{-4}
C _{in}	μg m ⁻³	0.26
C _p ⁱⁿ	μg g ⁻¹	2.2×10^3
F _{df}	g m ⁻² h ⁻¹	3.2×10^{-4}
C _{df}	μg g ⁻¹	1.0×10^3
M _{fl}	g m ⁻²	0.18 ^a
C _{fl}	μg g ⁻¹	2.8×10^2
TSP _{out}	g m ⁻³	6.4×10^{-5}
C _p ^{out}	μg g ⁻¹	6.4×10^3

^aFor coarse particles

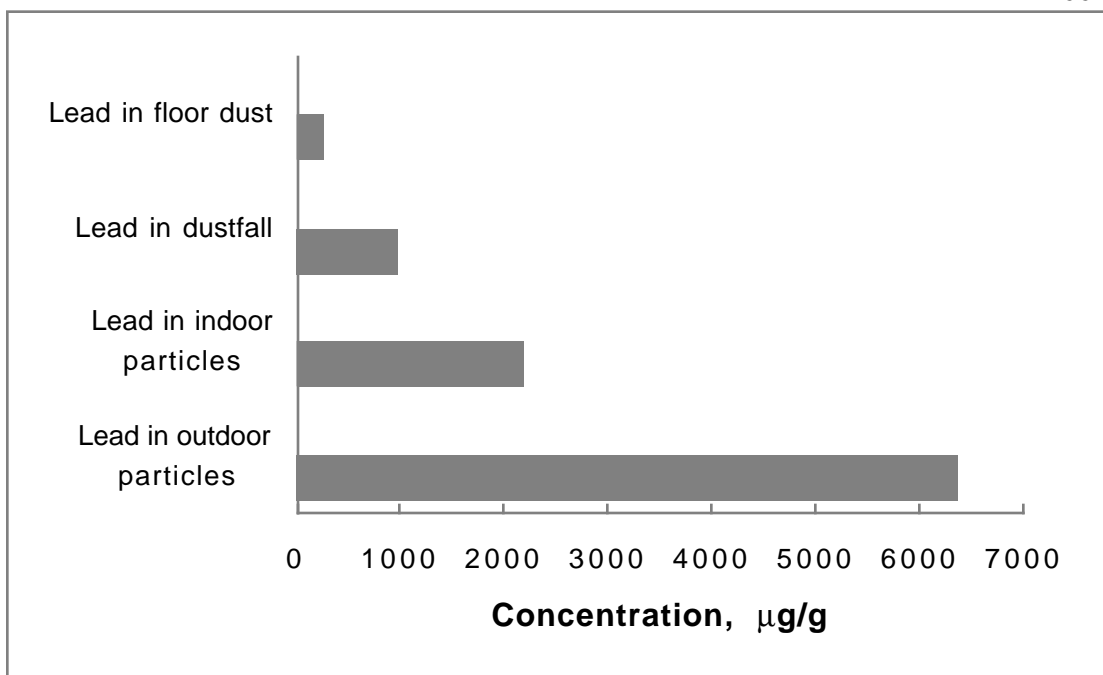


Figure 1. Concentrations of Pb in particulate matter sampled in the Arnhem Lead Study.

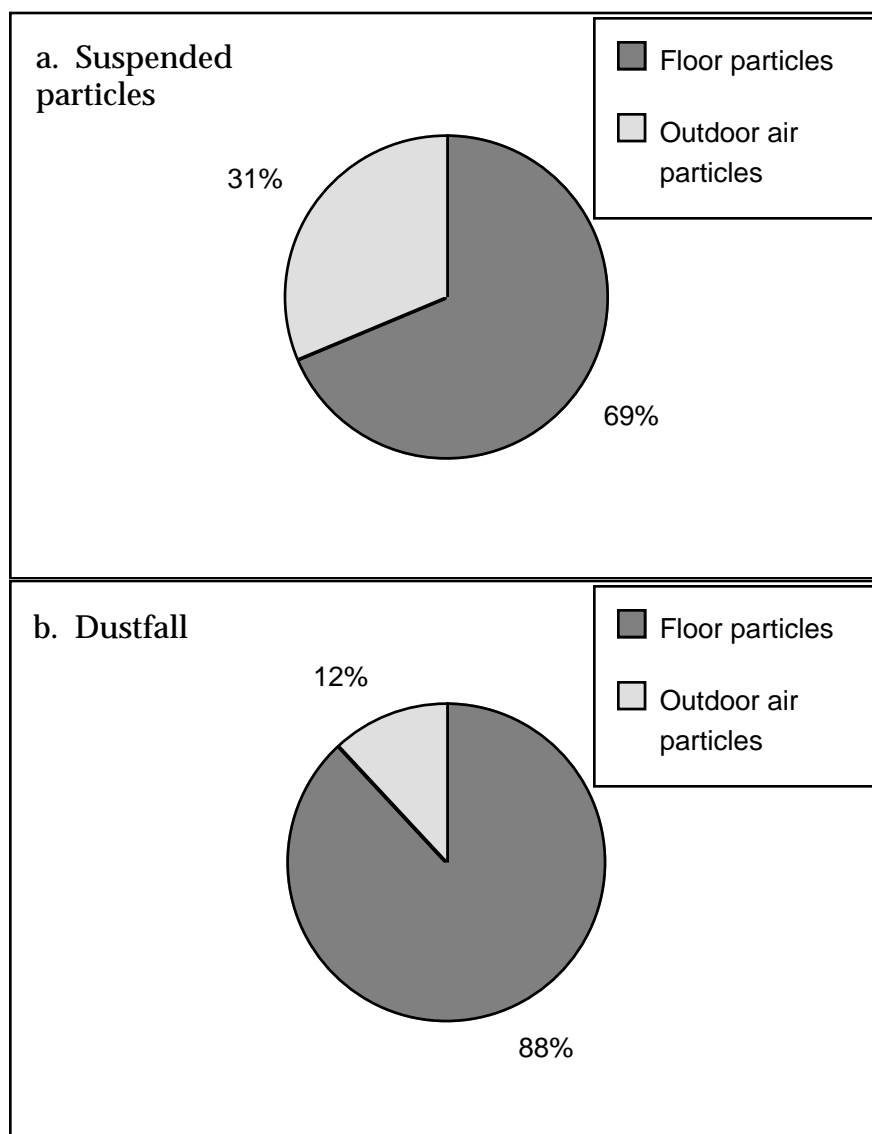


Figure 2. Sources of lead in suspended particles indoors and dustfall based on data from the Arnhem Lead Study.